CYCLING OF ^{137}CS IN SOIL AND VEGETATION OF A FLOOD PLAIN 30 YEARS AFTER INITIAL CONTAMINATION[a]

R. C. Dahlman and P. Van Voris[b]

Oak Ridge National Laboratory
Environmental Sciences Division
Oak Ridge, Tennessee 37830

Abstract

Distribution of radiocesium was determined in soil and vegetation components of a flood plain contaminated by Manhattan Project operations in 1944. Thirty years after contaminated waste effluents were deposited in a temporary holding basin, practically all the soil ^{137}Cs was still within 60 cm of the soil surface. Maximum ^{137}Cs concentrations occurred in the 12- to 22-cm horizon. Concentrations throughout the flood plain were variable; maximum levels of ^{137}Cs exceeded 20,000 pCi/g; intermediate levels of 5,000 to 20,000 pCi/g were encountered along the watercourse, and concentrations less than 5,000 pCi/g were found along the flood plain margins. Relative concentrations in soil, roots, and above-ground vegetation (expressed as ratios on a gram per gram basis) were 0.6 for root/soil, 0.05 for above-ground vegetation/soil, and 0.03 for above-ground vegetation/roots. Ratios ranged from 0.001 to 0.53 for all species, and average ratios for the 30-year post-contamination study showed that the relative ^{137}Cs distribution between plants and soil has not changed significantly from distributions reported 15 years ago

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[b]Graduate student in Ecology Program at the University of Tennessee.

(plant/soil ratio 0.05 vs 0.03 by Auerbach et al., 1959). The results also indicated that ratios were higher at low soil-^{137}Cs concentration. Thus, when soil and environmental conditions remain unchanged over a 30-year period, the relative concentration of ^{137}Cs between plants and soil does not appear to change significantly as a function of time.

Introduction

Radiocesium from fallout and from nuclear reactor wastes is an important contributor to radiation dose when humans are exposed to anthropogenic radioactivity. Maximum concentrations in foodstuffs and biota are usually coincident with incipient introduction to ecosystems (Russell, 1963; Ward et al., 1966; Porter et al., 1967; Wilson et al., 1969); once ^{137}Cs reacts with soil minerals, its availability to biota decreases substantially (Nishita et al., 1968; Waller and Olson, 1967). Exceptions, however, have been reported for highly weathered soils that contain a low abundance of micaceous minerals (Sharitz et al., 1975; Cummings et al., 1969) and for soils exhibiting low pH (Evans and Dekker, 1969).

One important question concerns the long-term geochemical fate of ^{137}Cs. As a result of time-dependent geochemical processes in soil, will the ^{137}Cs content of biota increase or decrease in the future? Or will the nuclide remain fixed to soil minerals as predicted by experimental studies (Tamura and Jacobs, 1960; Jacobs, 1960), and as determined by field observation after 10 to 20 years of ^{137}Cs contact with lacustrine deposits (Lomenick and Tamura, 1965). Geochemistry data suggest effective fixation by clay-type minerals (Fairbridge, 1972), and in a recent review it was concluded that present-day ^{137}Cs behavior in ecosystems is consistent with established facts about the biogeochemistry of the element (Dahlman et al., in press).

A site contaminated 30 years ago during Manhattan project operations provides an excellent opportunity for validating projections about the long-term fate of ^{137}Cs in ecosystems. The purpose of this paper is to report relative distributions of ^{137}Cs in soil and vegetation components of a flood-plain ecosystem contaminated in 1944. Results from a survey made in 1974, 30 years post-contamination, are compared with literature data obtained throughout the nuclear age and based on experimental and short-term studies on ^{137}Cs uptake by plants. The supposition that cesium is effectively fixed by soil clay (thus it is relatively unavailable to biological entities) is evaluated from this 30-year-old contamination event.

Methods and Materials

Radiocesium released from Manhattan Project Operations in 1944 was deposited in sediments of a temporary holding pond over a period of approximately 6 months. Other fission products (^{90}Sr, ^{106}Ru) and transuranic elements (^{239}Pu, ^{241}Am) were also deposited. As a result of dam failure in late 1944, the pond drained, and a young flood-plain forest has developed on the 2-ha. site since that time; maximum age of trees is 30 years. Relative concentrations of ^{137}Cs in soil, roots, and above-ground vegetation were determined in components of the flood-plain ecosystem.

Experimental Site

A typical flood-plain forest has developed on the previously flooded site. The three different forest communities contain common lowland species (ash, sycamore, boxelder, willow, sweet gum; Fig. 1). Understory and ground vegetation are representative of successional communities of flood plains in the Tennessee Valley. The fertile alluvial soils are representative of bottomlands; texture is predominantly sand and silt. Gravel lenses occur irregularly throughout the profile--probably related to historical meanderings of White Oak Creek. The creek presently bisects the flood plain, and although occasional flooding prevails, the creek has not overflowed during the past year.

Sampling Methods

A sampling grid was established on 30-m spacings over a 20,000 m^2 area of the flood plain. At each intersection point of the transects (Fig. 1), the above-ground portions of three different species of ground vegetation (*Microstegium vimineum*, *Impatiens capensis*, *Lonicera japonica*) were collected for radioanalysis. Foliage samples were collected in September 1974 before leaf fall. Samples were dried to a constant weight, shredded and homogenated, and subsamples of uniform geometry were analyzed for ^{137}Cs. Two dominant overstory trees were selected at each 30-m grid intersection. Leaf samples were collected from the species nearest the intersection; the second species sampled was in the opposite (180°) quadrant. Tree foliage was prepared for ^{137}Cs analysis in the same manner as for the ground vegetation collection. Radiocesium content of subsamples of uniform geometry was determined with a 3 x 3 in. NaI(Tl) well-type crystal connected to a Nuclear Data 812 pulse-height analyzer. Results were expressed in terms of picocuries per gram of oven dry weight.

Soil cores were taken near each transect intersection. Each core was located between each selected tree and the grid intersection at a point on the ground below the outer limit of the tree canopy. Each 7.5-cm-diam core was then sectioned into the following centimeter increments: 0-2, 2-5, 5-7, 7-12, 12-17, 17-22, 22-32, 32-42, 42-52, 52-bottom. Woody roots were manually removed from each increment; a random subsample of soil was taken from each increment for ^{137}Cs determination. The counting procedure was similar to that already described for vegetation; results were expressed in terms of picocuries per gram of oven-dry (100°C) soil.

Roots were cleaned using an ultrasonic bath. The procedure was uniformly effective, and all roots received the same treatment. When selected samples were examined microscopically, minute quantities of soil were observed adhering to the root surface. Thus, roots were not absolutely clean. A measure of ^{137}Cs before and after cleaning in the ultrasonic bath revealed a 66% ± 0.94[a] reduction in activity. The low coefficient of variation (CV = 25%) of the percentage ^{137}Cs removed by cleaning indicated the uniformity of treatment. Cleaned roots were radioassayed according to procedures described for foliage, and the results were expressed in terms of picocuries per gram of oven-dry root mass.

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[a]Plus or minus 1 standard error of mean.

Results

Radiocesium in all soil cores collected from the flood plain exceeded background concentrations (3-6 pCi/g from fallout contamination). Distribution patterns show the highest concentrations along White Oak Creek (5,000 to 20,000 pCi/g), near the dam and at the upper portions of the flood plain (greater than 20,000 pCi/g) (Figs. 2,3). Concentrations were less than 5000 pCi/g along the lateral margins of the flood plain.

Patterns of depth distribution of the ^{137}Cs are given in Figs. 2a, 2b, 2c, and 2d for 0-12, 12-22, 22-42, and 42-62 cm increments respectively. In terms of both soil concentrations and total area contaminated, ^{137}Cs was most prevalent in the 12-22 cm horizon; the maximum area possessing greater than 20,000 pCi/g occurred at this depth. Radiocesium greater than 100 pCi/g was found in only 10 cores at depths > 42 cm; however, when present below 42 cm, the concentration usually exceeded 5000 pCi/g (Fig. 2d). Concentrations were summarized as weighted averages for the entire profile, and an integrated ^{137}Cs value was calculated for the entire profile (Fig. 3).

The distribution pattern of mean root concentrations was related to the pattern of ^{137}Cs in soil (Fig. 4). Radiocesium concentration in roots ranged from 100 to 3000 pCi/g, and root/soil ratios of radiocesium concentration ranged from 0.04 to 3.6 ($\bar{X}$ = 0.60 ± 0.04). Maximum concentrations in roots were observed in samples collected near the dam and towards the upper portion of the flood plain. Intermediate concentrations were observed along White Oak Creek, and the lowest values were on lateral margins of the flood plain. However, when ratios of ^{137}Cs

concentration in roots to concentration in soil were calculated, the highest values exhibited a different distribution pattern (Fig. 5). Ratios greater than 0.25 were evident along the margins and along the S 240 transect across the center of the flood plain. Near the dam and at the upper portion of the flood plain the ratios were consistently less than 0.25. Although ^{137}Cs concentration in roots generally was proportional to soil concentration, the distribution pattern of ratios indicated that roots collected from the shaded zone of Fig. 5 contained more ^{137}Cs relative to the soil concentration, resulting in higher ratios.

Average radiocesium concentrations in above-ground vegetation ranged from 2 to 160 pCi/g for tree species and from 16 to 108 pCi/g for ground-cover species. Because the ^{137}Cs level of above-ground vegetation generally corresponded to patterns of soil concentration, the results are expressed in terms of relative concentration; viz., ratios of $[^{137}Cs]_{veg}/[^{137}Cs]_{soil}$. Ratios ranged from 0.001 to 0.23 for overstory species and from 0.001 to 0.53 for ground-cover species. The average ratio was 0.05 for overstory and ground-cover species.

For ground vegetation, the distribution pattern (Fig. 6) of the $[^{137}Cs]_{veg}/[^{137}Cs]_{soil}$ ratio was similar to the pattern observed for roots. Highest ratios for ground-cover were observed along the flood-plain margins; i.e., the locations where soil-^{137}Cs concentration were lowest. A similar but less distinct pattern of ratio distribution occurred (not illustrated) for overstory species. Again, the highest ratios were observed in areas of the flood plain where soil concentrations were lowest.

Discussion

Radiocesium in all components (soil, root, foliage) of the flood-plain ecosystem is attributed to historical releases during the Manhattan Project. Highest concentrations appear along the diverted watercourse (currently the new channel for White Oak Creek) and in the deeper sediments next to the old dike. Radiocesium and associated sediments were apparently deposited along a watercourse due to diminished water velocity as White Oak Creek entered the temporary pond in 1944. It is possible that the entire contaminated area has not yet been completely identified, because cores possessing the highest level of ^{137}Cs were collected on the S 120 transect outside the present estimated pond boundary. Evidently, the temporary pond extended further upstream than is indicated by the present boundary.

Total quantity of ^{137}Cs in the flood-plain soil is estimated to be 105 Ci[b] based on an integration of concentration data given in Figs. 2a, 2b, 2c, and 2d over the 0 to 62 cm depth. Except for parts of the

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[b]Total ^{137}Cs budget (B) in soil is calculated according to $B = \Sigma I_a$, where I_a, the activity increments, are $I_{2,500}$, $I_{12,500}$, and $I_{25,000}$; i.e., estimated mean concentrations for gradients given in Figs. 2a, 2b, 2c, and 2d. For example,

$$I_{2,500} = \frac{(6.09 \times 10^3\ m^3)\ (10^6\ cm^3\ m^{-3})\ (1.4\ g\ cm^{-3})\ (2.5 \times 10^2\ pCi\ g^{-1})}{10^{12}\ pCi\ Ci^{-1}}$$

$$= 21.4\ Ci.$$

flood plain inadvertently omitted from the 1974 survey, this total represents the entire present budget of ^{137}Cs in soil because essentially no activity was observed below the 62 cm depth. Thus, there has not been extensive migration of ^{137}Cs to deeper horizons of the flood plain over the 30-year period since initial deposition. This observation confirms earlier predictions (Jacobs, 1960) that ^{137}Cs would be effectively fixed by the illitic clay component of Oak Ridge soils.

Within the 60-cm zone, however, the results indicate possible minor ^{137}Cs redistribution throughout the soil profile because without exception the highest ^{137}Cs concentration of each core was found below the surface horizon. On the average, maximum ^{137}Cs was observed in the 12-22 cm horizon (mean depth of 14 cm for entire flood plain), Fig. 2b. It is difficult to explain how these depth distributions became established in the absence of better documentation of stratification of the initial deposit, early sedimentation rates, and recent erosion in relation to flooding and meandering of White Oak Creek. A Tennessee Valley Authority (1951) survey of White Oak Creek estimated sedimentation equivalent to a depth of 9 cm--if sedimentation occurred uniformly over the entire flood plain. No data are available on sedimentation or erosion since that time. The 9-cm layer of sediment, if deposited uniformly on top of the 1944 input of ^{137}Cs, would be a partial explanation for the maximum concentration observed at an average depth of 14 cm. Since ^{137}Cs maxima of a number of cores were observed below 22 cm, it would appear that, in addition to unequal sedimentation on the flood plain, other mechanisms may also be responsible for the observed vertical distributions in the soil. A second possible mechanism is a combination of diffusion-connection as rainfall percolates through the soil profile. We have not made calculations of potential migration using diffusion coefficients and K_d's for the flood plain soil. A third mechanism could involve burrowing and mixing activities of soil fauna. Reichle et al. (1971) showed that earthworms can cause complete turnover of soil to a 75 cm depth in 60 years. Both earthworms and soil-inhabiting crayfish are common at the flood plain site.

Increased ^{137}Cs in subsoil horizon contrasts somewhat with the depth distribution reported for White Oak Lake sediments in mid-1960 (Lomenick and Tamura, 1965). They reported a maximum concentration in the 0-6 in. (0-15 cm) depth for lacustrine sediments of the lake compared with ^{137}Cs maxima below 22 cm for the flood plain. Although minor differences are evident for distributions of ^{137}Cs between White Oak Lake and for the flood plain reported herein, major vertical patterns are similar. Negligible ^{137}Cs was found below 60 cm at both sites.

Radionuclide concentrations for roots are rarely reported because of the difficulty of collecting a representative root sample and because contaminated soil is not easily removed from roots. We recognize that such limitations would be incumbent on our observed ^{137}Cs concentrations for roots. However, we observed appreciable removal of soil-^{137}Cs from roots by the sonic washing technique (66% ± 0.94 of the initial activity). Consistency of cleaning is illustrated by the relatively low coefficient of variation (CV = 25%). By this technique, we also assumed that leaching losses due to washing were approximately compensated by increased root ^{137}Cs due to residual soil contamination. Although the relative magnitude of either error is impossible to estimate, the data are considered sufficiently adequate for qualified conclusions about ^{137}Cs relationships involving soil, roots, and above-ground vegetation.

The concentrations of ^{137}Cs in roots were related to soil concentrations of ^{137}Cs. Such a relationship is to be expected according to concepts of passive uptake of non-essential trace elements by plant roots; viz., root concentration is proportional to element concentration of the rhizosphere. Root/soil ratios of ^{137}Cs suggest another interesting pattern; the higher ratios (greater than 0.25) were associated with soil possessing relatively low ^{137}Cs concentrations (compare the high ratio of the shaded area of Fig. 5 with the less than 5,000 pCi/g zone of Fig. 3). A similar relationship has been observed for plants growing on a ^{137}Cs-contaminated flood plain at Savannah River, South Carolina (Sharitz et al., 1975). Root-soil ratios were a factor of 28 higher for plants growing in "low-level" soil (26 pCi/g) compared with plants in "high-level" soil (545 pCi/g).

The variable ratios observed in both studies (Oak Ridge and Savannah River) are probably related to soil factors that regulate ^{137}Cs sorption by clays or inversely the availability to roots as governed by different exchange characteristics of endemic clays. But in the absence of more definitive experimental data the exact mechanism responsible remains unexplained. Because the inverse relationship between ^{137}Cs concentrations in roots and soil appears to be a common phenomenon in aged deposits at Savannah River and Oak Ridge, the relationship needs further interpretation in the context of long-term hazards evaluation. Variable plant uptake, as reflected by the factor of 10 to 100 difference in ratios (which in turn appears to be dependent on ^{137}Cs concentration in soil), illustrate, again, that constant coefficients of plant uptake should be used cautiously, if at all, in generic assessments of radiologic hazard.

Results from ^{137}Cs measurements of both roots and above-ground vegetation clearly illustrate the differential concentrations in plants growing on the contaminated flood plain. Concentrations in roots were 10 to 100 times greater than ^{137}Cs for above-ground vegetation. Although some ^{137}Cs in roots may be attributed to soil residue, the fact that concentrations in roots were different from tops by at least an order of magnitude indicate <u>in situ</u> retention of root-absorbed ^{137}Cs. On a concentration ratio basis, only 1 to 10% of the root-^{137}Cs appears to reach above-ground parts.

The relative concentration of ^{137}Cs between above-ground vegetation and soil is an indicator of the nuclide's availability to biotic components of ecosystems. Concentration ratios averaged 0.05 for both overstory and ground-cover species. The inverse relationship of ^{137}Cs concentrations in above-ground vegetation vs concentrations in soil were again manifest but less markedly than for roots vs soil. For example, ratios exceeding 0.02 were evident for zones in which soil concentration was less than 5,000 pCi/g while ratios were less than 0.02 where ^{137}Cs in soil exceeded 5,000 pCi/g.

Because the vegetation/soil ratio can be used as an indicator of ^{137}Cs uptake in relation to temporal and site characteristics, the average ratio for this site is compared with ratios derived from other experiments and ecosystem studies (Table 1). The usual ratio is less than 1.0; for most agricultural soils, it is much less than 1.0. High values, e.g., ratios of 10 to 20 reported by Sharitz (1975) and Cummings (1969), are probably related to the absence of 2:1 layered silicates which tend to fix Cs irreversably as described by Dahlman et al. (in press). The low ratios reported herein (0.001 to 0.53) confirm that ^{137}Cs is relatively immobile in terrestrial ecosystems at Oak Ridge. These results tend to confirm earlier projections (Tamura and Jacobs, 1960; Lomenick and Tamura, 1965) that ^{137}Cs would be effectively fixed and retained by soil minerals.

The most striking result obtained from the flood plain study is that the average vegetation/soil ratio for ^{137}Cs (0.05) is nearly the same as earlier values (0.03) reported by Auerbach et al. (1959,1972) for similar lacustrine deposits (White Oak Lake sediments). That ratios are nearly equivalent for earlier short-term cropping experiments and for a forest ecosystem growing for 30 years on a contaminated site is an important confirmation of the established concept about the biogeochemical fate of radiocesium. The ratio remained relatively constant for different treatment and temporal conditions (field crop, greenhouse pot culture, natural ecosystem). Over the 30-year period, it appears on the average that radiocesium concentrations in plants have not been influenced by the time factor of soil and ecosystem processes. When soil and environmental conditions remain relatively constant, relative uptake of ^{137}Cs by plants does not appear to change significantly as a function of time; average ratios remain relatively constant. Yet appreciable variation in plant-soil ratios (0.001 to 0.53) suggests that ^{137}Cs uptake is not a constant function of soil concentration; ratios are higher when [^{137}Cs] of soil is lower.

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Table 1. Relative Concentration of ^{137}Cs in Plants and Soil

Plant Species or Site	Soil and/or Experimental Characteristics	Ratio	Reference
Oak Ridge Flood plain Ecosystem	Alluvial deposit		
	Overstory species	Range 0.001 to 0.23	This report
	Ground cover species	Range 0.001 to 0.53	
	Grand mean	0.05	
Savannah River Flood plain; (*Sagittaria latifolia*, *Polygonum punctatum*)	Alluvial deposit; mean for two species and soil concentrations		
	"low-level"	10.8 to 20.1	Sharitz et al. (1975)
	"high level"	0.6	
White Oak Lake Bed; millet	Field plots on lacustrine deposit	0.03	Auerbach et al. (1959)
White Oak Lake Bed; millet	Continuous cropping of lake-bed soil in green-house pots	0.03	Auerbach et al. (1972)
Beans	pH 6.8; typical agri-cultural crop soil	0.013 to 0.027	Nishita et al. (1968)
Oats	Soils from across the southeastern United States	0.02 to 72	Cummings et al. (1969)

Figure Explanation--Fig. 1

Flood Plain Perimeter	————
Area not studied	X X X X
White Oak Creek and flood channel	-..-..-
Drainages	-...-...-
Boundary of principal communities	————
Boundary of minor communities (understory and ground cover)	
Dogwood	....
Wild rye	---------
Cattails	-.-.-.-.-.

Description of principal communities:

1. Open area (no overstory trees) with scattered dogwood (*Cornus amomum* L.), boxelder (*Acer negundo* L.), willow (*Salix nigra* Marsh), walnut (*Juglans nigra* L.), and white ash (*Fraxinus americana* L.). Ground cover consists of jewel weed (*Impatiens capensis* L.), microstegium (*Microstegium vimineum* Nees), aster (*Aster sp.*), and golden rod (*Solidago sp.*).

2. Stand dominated by sycamore (*Plantanus occidentalis* L.), ($\bar{X}$ = 3 trees/100 m^2). Second dominant tree is white ash (*Fraxinus americana* L.).

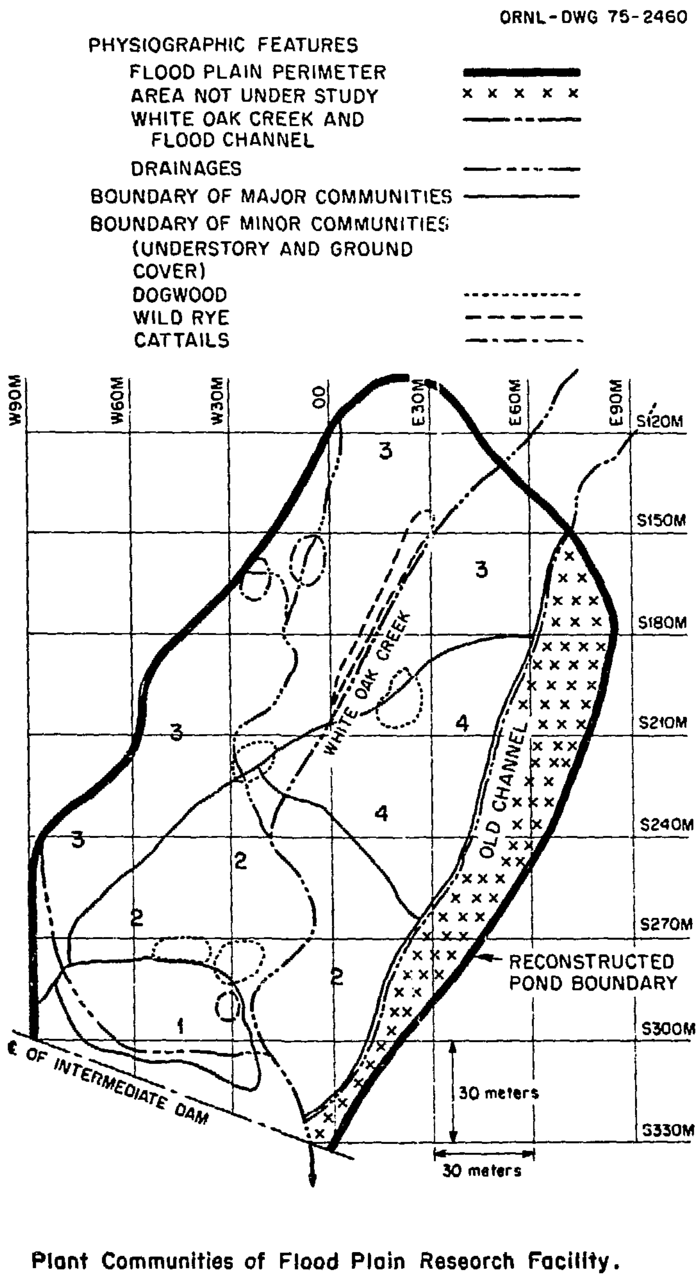
3. Stand dominated by white ash (*Fraxinus americana* L.), ($\bar{X}$ = 8 trees/100 m^2); associated overstory species of sycamore (*Plantanus occidentalis* L.), black willow (*Salix nigra* Marsh), red maple (*Acer rubrum* L.) boxelder (*Acer negundo* L.), tulip poplar (*Liriodendron tulipifera* L.), sweet gum (*Liquidambar styraciflua* L.), and elm (*Ulmus americana* L.).

4. Mixed hardwood region with no single dominant overstory species, co-dominance of elm (*Ulmus americana* L.), and tulip-poplar (*Liriodendron tulipifera* L.); with associated overstory of red cedar (*Junipirus virginiana* L.), black willow (*Salix nigra* Marsh), red maple (*Acer rubrum* L.), white ash (*Fraxinus americana* L.), sycamore (*Plantanus occidentalis* L.), sweet gum (*Liquidambar styraciflua* L.), boxelder (*Acer negundo* L.), walnut (*Juglans nigra* L.), and hackberry (*Celtis occidentalis* L.). The principal understory species are dogwood (*Cornus amomum* L.), white ash (*Fraxinus americana* L.), grape (*Vitis sp.*), plum (*Prunus sp.*), buckeye (*Aesculus Octandra Marsh*), beech (*Fagus grandifolia* Ehrh.), and magnolia (*Magnolia acuminata* L.).

Except where a dense, homogenous stand is noted, the ground cover consists of jewel weed (*Impatiens capensis* L.), microstegium (*Microstegium vimineum* Nees), aster (*Aster sp.*), golden rod (*Solidago sp.*), trumpet vine (*Campsis radicans* Lour.), Japanese honeysuckle (*Lonicera japonica* L.), vetch (*Vicia sp.*), poison ivy (*Rhus radicans* L.), sedge (*Carex sp.*), and bull rush (*Scirpus sp.*).

Scattered understory species through entire area are dogwood (*Cornus amomum* L.), boxelder (*Acer negundo* L.), sweet gum (*Liquidambar styraciflua* L.), willow (*Salix nigra* Marsh), walnut (*Juglans nigra* L.), plum (*Prunus sp.*), and buckeye (*Aesculus Octandra Marsh*).

Widespread shrub species throughout the entire area include wild rose (*Rosa setigera* L.), blackberry (*Rubus sp.*), alder (*Alnus sp.*), elderberry (*Sambucus canadensis* L.), and grape (*Vitis sp.*).



Plant Communities of Flood Plain Research Facility.

Fig. 2. Distribution of ^{137}Cs (pCi/g) in different soil depths of the flood plain. Concentrations at each grid intersection are an average of two cores. Weighted average is based on measurements for subincrements of each depth:

Fig. 2a. 0 to 12 cm, 3 increments; 0 to 2, 2 to 7, and 7-12 cm.

Fig. 2b. 12 to 22 cm, 2 increments; 12 to 17 and 17 to 22 cm.

Fig. 2c. 22 to 42 cm, 2 increments; 22 to 32 and 32 to 42 cm.

Fig. 2d. 42 to 62 cm, 2 increments; 42 to 52 and 52 to 62 cm.

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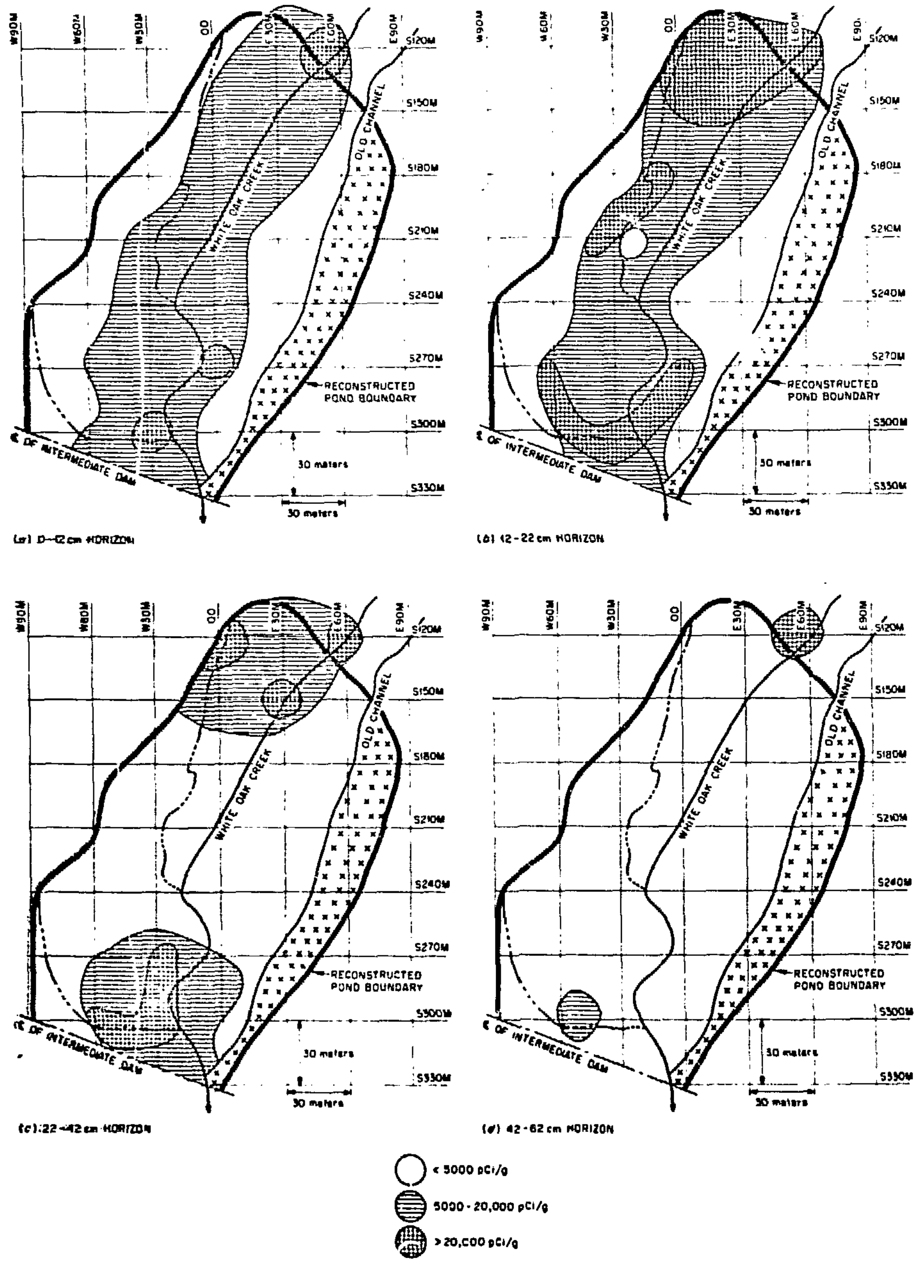

Fig. 3. Weighted average of ^{137}Cs (pCi/g) for entire 62-cm soil profile.

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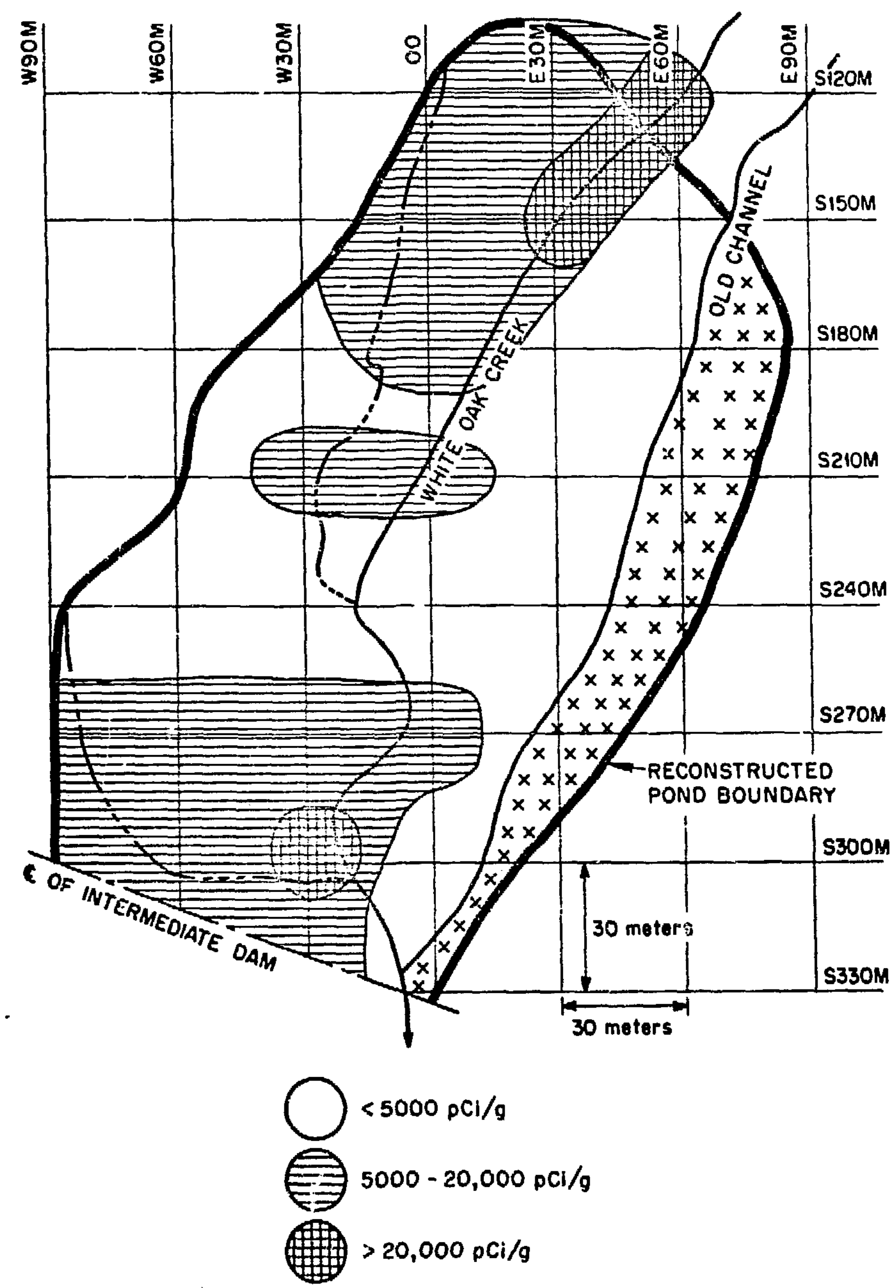

Soil Core Weighted Averages.

Fig. 4. Mean root ^{137}Cs (pCi/g) for flood plain.

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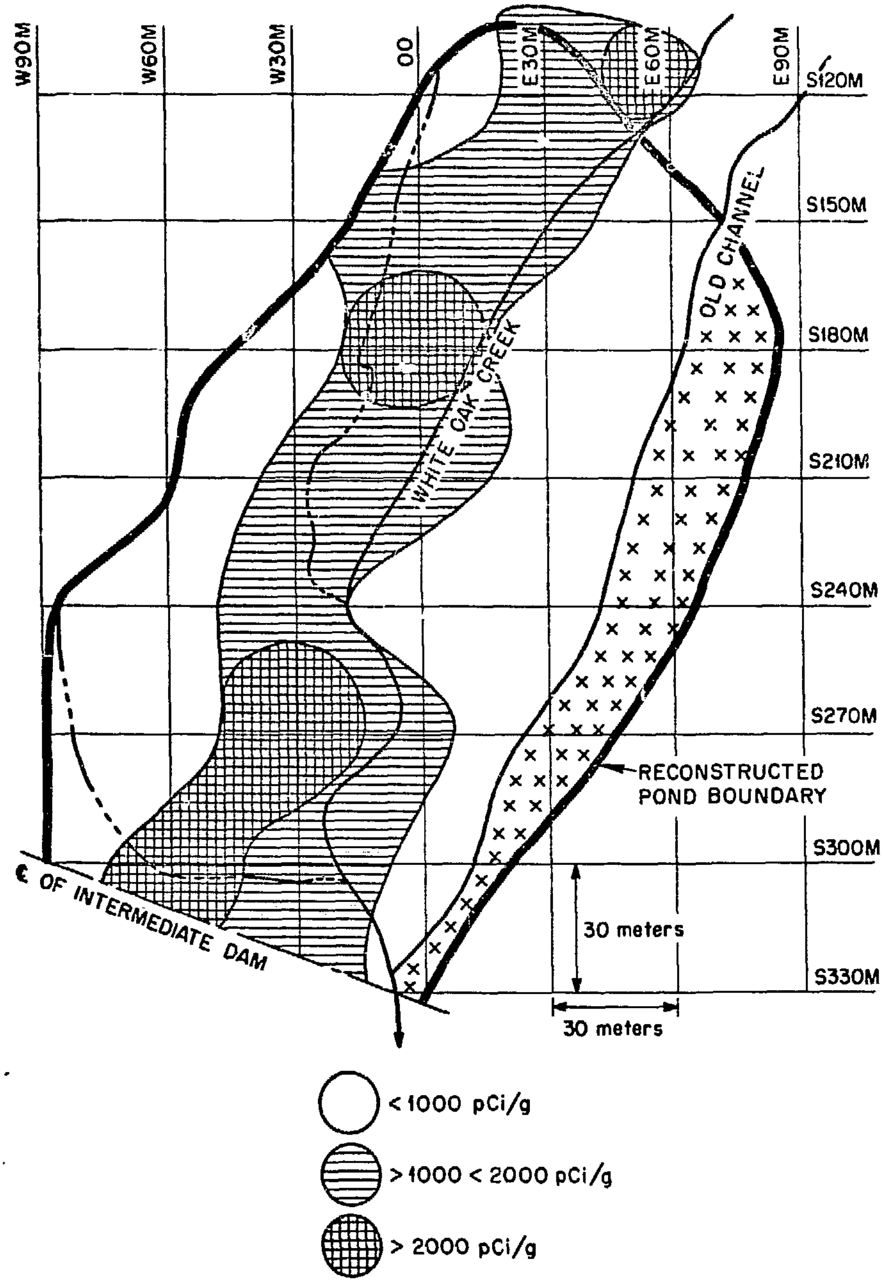
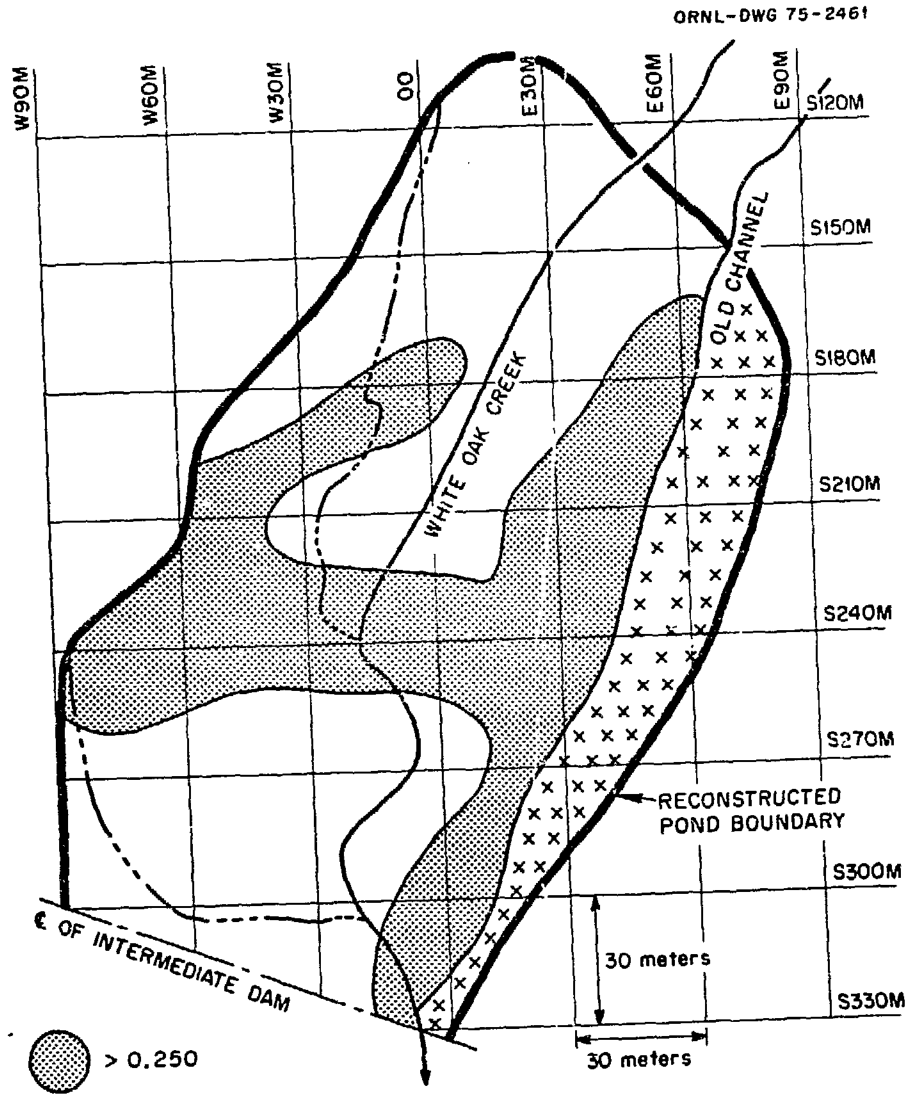


Mean Root ^{137}Cs pCi/g.

Fig. 5. Distribution of root/soil concentration ratios: $[^{137}Cs]_{root}/[^{137}Cs]_{soil}$ on gram-per-gram basis.



$\frac{\text{ROOTS}}{\text{SOIL}}$ RATIO

Fig. 6. Distribution of ground vegetation/soil concentration ratios: $[^{137}Cs]_{vegetation}/[^{137}Cs]_{soil}$ on a gram-per-gram basis. Ground vegetation sampled were Microstegium vimineum, Impatiens capensis, and Lonicera japonica.

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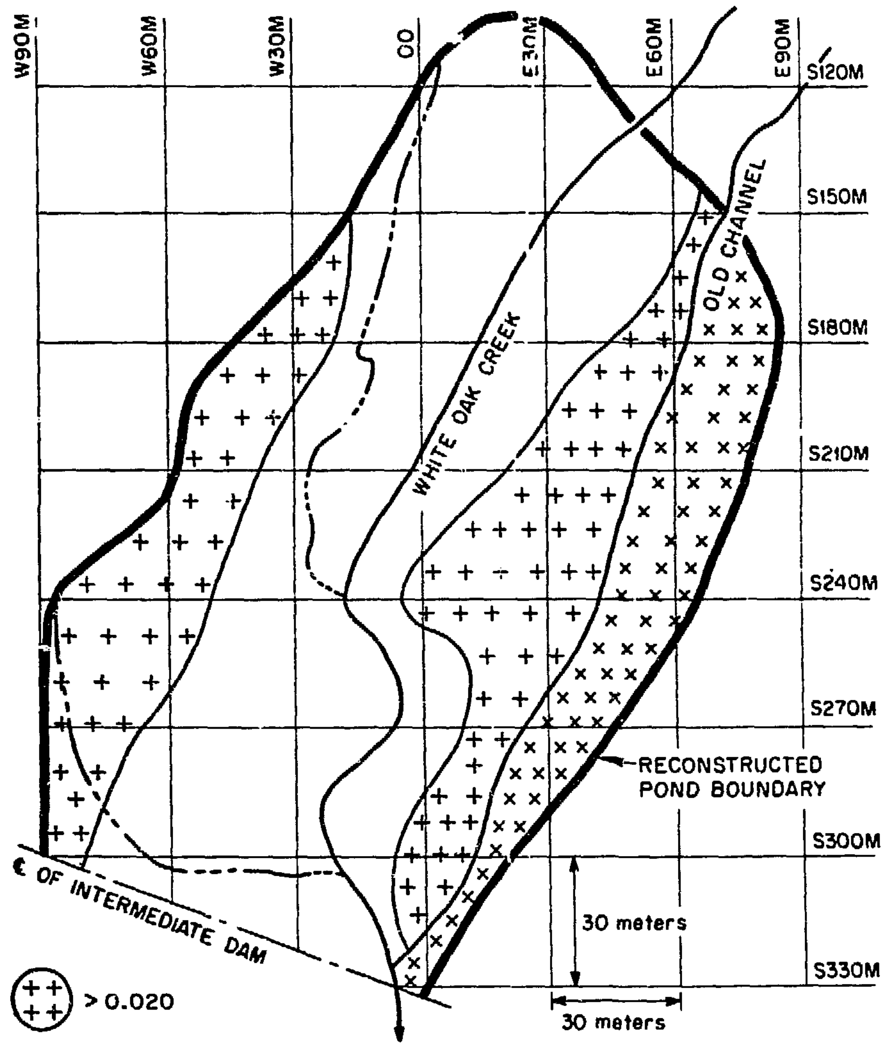

GROUND VEGETATION / SOIL RATIO